

REMARKS

By this Preliminary Amendment, the Specification has been amended to include a cross-reference to related applications which has been inserted in page 1. Also pages 1, 5, and 10 have been amended to recite the Specification section headings required by U.S. practice. In addition, pages 4 and 5 of the U.S. Specification have been amended to conform to the changes made in the International Office.

The amendments to the claims are to cancel the originally filed claims 1 to 22 without prejudice, and to replace these claims with new claims 23 to 43. New claims 23 to 43 eliminate the multiple dependency of the claims, so as to avoid the U.S.P.T.O. surcharge therefor. Also claims 23 to 43 have been written so as to comply with U.S. formal requirements. Claims 23 to 43 correspond to the amended claims filed in the International Office.

An Abstract of the Disclosure on its own separate page is enclosed.

No new matter has been introduced by this amendment. Entry of this amendment is respectfully requested.

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MARKED-UP VERSION
OF
AMENDED SPECIFICATION

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Process for producing monolithic oxidation catalysts
and their use in the gas-phase oxidation of
hydrocarbons

CROSS REFERENCE TO RELATED APPLICATIONSBACKGROUND OF THE INVENTION

5 1. Field of the Invention
The invention relates to a process for
producing monolithic oxidation catalysts and to their
7 use in the gas-phase oxidation of hydrocarbons.

8 2. The Prior Art
Supported catalysts for the gas-phase oxidation
of hydrocarbons to give the corresponding oxidation
10 products such as carboxylic acids, carboxylic
anhydrides or aldehydes, which catalysts have a
catalytically active surface coating consisting
essentially of titanium dioxide (TiO_2) and divanadium
pentoxide (V_2O_5), have been known for a long time. A
15 typical example of the use of such catalysts is the
preparation of phthalic anhydride, in which mixtures of
o-xylene and air or naphthalene and air or o-xylene,
naphthalene and air are passed over an appropriate
catalyst in a shell-and-tube reactor. The heat
20 generated in this strongly exothermic reaction is
customarily removed (cooling, isothermal reaction
conditions) by means of a salt melt which surrounds the
reaction tubes.

The supported catalysts used here comprise an
25 inert support body, for example having a ring shape or
a spherical shape, on which the actual catalytically
active composition is present. The active composition
consists predominantly of the main components TiO_2 in
the anatase form and V_2O_5 . To improve the control of the
30 activity and to improve the selectivity, further
activating or deactivating additives, for example
oxides of transition elements or alkali metal
compounds, are frequently added in small amounts as
dopants (promoters) to the catalytically active
35 composition.

The supported catalysts are generally produced
by spraying aqueous suspensions or aqueous solutions of
 TiO_2 and V_2O_5 , frequently with addition of promoters and

downstream adiabatic reactor. This process variant is likewise intended to enable the phthalic anhydride formed to be obtained largely free of by-products and without a loss in yield. Here too, a shaft oven is claimed as adiabatic reactor.

6 AT-A 9201926 describes a process for preparing phthalic anhydride in a main reactor having a downstream adiabatic reaction zone. Here, it is expressly stated that, for economic reasons, it is advantageous for the adiabatic reactor to be connected directly to the main reactor. Apart from the use of catalyst base bodies in the form of rings or spheres, mention is also made of the possible use of a catalyst having a monolithic structure, for example in the form of a honeycomb. In the reaction procedure described, this measure enables the yield of phthalic anhydride to be increased by about 1% by weight. The formation of the by-product phthalide is at the same time significantly reduced. The use of the monolithic catalyst shape mentioned is not described in any example. Likewise, no details are given of the production of a monolithic catalyst and associated problems.

23 Owing to the laminar flow occurring in honeycomb catalysts, they have only a very low pressure drop even at very high gas velocities. However, a disadvantage is that, owing to the lack of turbulent flow resulting from the shape, heat and mass transfer in the honeycomb channels, and thus heat removal, are greatly reduced. This situation makes use of honeycomb catalysts as catalyst supports virtually impossible for strongly exothermic processes in conjunction with a selective oxidation. Honeycomb catalysts have therefore become established industrially only in waste gas purification or waste gas incineration where all the organic constituents undergo total oxidation to CO₂.

Coating monolithic support material with a catalytically active composition comprising the main

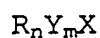
constituents TiO_2 , V_2O_5 and possibly dopants by generally known methods, for example a dipping process, is found to be impractical. This is because coating suspensions based on commercially available TiO_2 have a
5 very high viscosity even at solids concentrations of 30-35% by weight and thus make coating of the channels of a monolithic support material virtually impossible without blocking the channels.

In order to coat monolithic catalyst supports
10 with the necessary amount of catalytically active composition, for example 50-150 g of active composition per liter of catalyst, the coating process would have to be carried out with such a low-concentration "active composition" suspension that the necessary layer
15 thickness would be achieved only after repeating the coating process a number of times. However, this at the same time once again increases the problem of blocking of the channels in the catalyst support because of the multiple coating steps. Furthermore, this is associated
20 with significantly more work and thus with increased costs and is therefore uneconomical.

21 ~~SUMMARY OF THE INVENTION~~
22 It is therefore an object of the invention to provide a simple and preferably single-stage process for producing monolithic catalysts based on TiO_2 /metal
25 oxides.

It has now surprisingly been found that the viscosity of highly concentrated TiO_2 coating suspensions having a high solids content can be greatly reduced by addition of surfactants.

30 The invention provides a process for producing monolithic supported catalysts for gas-phase oxidation by coating the catalyst support by means of a suspension, wherein the latter comprises catalytically active composition ^{and} comprising one or more types of TiO_2
35 and 1-10% by weight of one or more surfactants of the formula



The monolithic catalysts produced according to the invention are particularly suitable for preparing phthalic anhydride in an adiabatic reactor (after-reactor) in combination with an isothermally operated reactor (main reactor, for example filled with a bed of particulate catalyst).

The adiabatic reactor can also be operated advantageously with upstream gas cooling. In a particularly preferred embodiment, the upstream gas cooling and the adiabatic reaction are carried out in a joint apparatus.

In industry, it is customary to cool the reaction gas in a gas cooler before isolation of the product. The upstream gas cooling, the adiabatic reaction in the monolithic catalyst bed and further cooling can be carried out within the reactor or outside the reactor, or in a joint apparatus.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS
The invention is illustrated in more detail by the following examples.

The following examples demonstrate the influence of surfactants from the group consisting of phosphoric esters on the viscosity of support oxide suspensions. The flow behavior of the suspensions prepared was determined by a method based on DIN 53211 using a flow cup. Two types of TiO_2 which differed from one another only in the particle size were tested. The mean particle diameter was 0.1 and 0.4 μm , respectively. This measurement method was selected since it could appropriately simulate flow of a suspension out of the honeycomb channels. As outflow nozzle, use was made of a nozzle having a diameter of 2 mm corresponding to a flow orifice area of 3.14 mm^2 . For comparison, a 200 csi honeycomb has a channel cross section of 2.3 mm^2 and a 100 csi honeycomb has a channel cross section of 4.66 mm^2 . If the viscosity of the suspension was too high, i.e. the suspension could not flow through the nozzle under these conditions, the experiment was repeated using a nozzle opening of 4 mm